OVERVIEW OF FLAMMABLE GAS GENERATION RESEARCH IN THE DOE COMMUNITY FOR FISCAL YEAR 1998

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1.0 INTRODUCTION

The radiolytic production of flammable gases in hydrogenous materials containing radioactive matter can pose a safety hazard in both transportation and storage. The resolution of the issues involving flammable gas generation affects national programs related to transportation and storage of contact and remote-handled transuranic waste, low-level waste, weapons grade plutonium and other radioactive materials shipped or stored in the DOE complex.

In support of the National Transportation Program, Waste Management Federal Services Northwest Operations (WMNW) conducted an overview of hydrogen gas generation research underway in the DOE complex. The overviews included visiting laboratories at Rocky Flats, Idaho Falls, and Los Alamos to view and discuss work in progress. The purpose of this report is to provide a synopsis of the ongoing experimental work.

2.0 BACKGROUND

Currently Lawrence Livermore National Laboratory (LLNL) is preparing a NUREG for the U.S. Nuclear Regulatory Commission (NRC) which will provide guidance on the transportation and storage of packages containing payloads which radiolytically produce hydrogen gas. The NUREG publication date is targeted at December 1999. Historically, the NRC IE Information Notice No. 84-72 has identified applicable flammable gas criteria for transportation. Based on the notice, hydrogen gas generation in a package is limited to the rate which will produce a molar quantity of 5% by volume in the secondary container gas void at STP in a period of time that is twice the expected shipment time. IE Information Notice No. 84-72 requires determination of hydrogen gas volumes by tests or measurements. However, historically the NRC has accepted analytical proof in lieu of testing.

In the absence of specific guidance, NRC and DOE safety analysis reports for certified packagings (SARs) have been required to undertake a burden of proof for hydrogen gas generation. SARs have been required to define and defend the analytical tools used to calculate hydrogen gas generation. Input parameters and assumptions have required extensive defense and, in most cases, bounding values and assumptions have been used which have resulted in overly conservative limits for shipment payloads and associated increased costs and schedule delays. Guidance on such issues as inerting, gas getters and recombiners, and lower flammability limits of mixtures of flammable gases has not been available. In addition, if a package does exceed 5% hydrogen gas by volume, there is no approved method of showing that a package provides an acceptable margin of safety and therefore meets the DOT requirements for containment.

Likewise, there is little specific guidance available for evaluation of gas generation in plutonium-bearing materials. DOE Standard 3013 *Criteria for Preparing and Packaging*

Plutonium Metals and Oxides for Long-Term Storage provides criteria for the packaging and storage of plutonium metals and stabilized oxides. However, the standard does not deal with other forms of plutonium or wastes containing fissile materials.

3.0 THE GAS GENERATION TEST PROGRAM AND THE MATRIX DEPLETION PROGRAM

A number of experimental programs are currently in progress within the DOE community. By far the largest body of work is being conducted as part of the payload expansion plan for the Transuranic Package Transporter-II (TRUPACT-II) shipments of contact-handled TRU waste to the Waste Isolation Pilot Plant (WIPP). Projects which support this effort include the Matrix Depletion Program (MDP) and the Gas Generation Test Program (GGTP) at the Rocky Flats Environmental Technology Site (RFETS) and at Idaho National Engineering and Environmental Laboratory (INEEL). The results of the program will be used to submit an application for NRC review with the object of increasing the allowable TRUPACT-II payload.

Both the GGTP and the MDP are outlined in detail in the following documents: *TRUPACT-II Payload Expansion Plan*, IT Corporation 1997; *TRUPACT-II Matrix Depletion Program Status Report Fiscal Year 1997*, Benchmark Environmental Corporation 1997. A draft of the TRUPACT-II Matrix Depletion Program status report for fiscal year 1998 not yet available at the end of September 1998.

The GGTP at RFETS and INEEL consists of controlled tests of Contact-Handled (CH) TRU waste containers to quantify the gas generation properties under simulated transportation thermal environments. CH TRU drums fall into two broad categories depending on their gas generation potential: test category and analytical category. A drum category is determined by waste type and wattage limit resulting from decay heat and is based on theoretical worst case calculations for gas generation. Test category drums require further testing for gas generation before acceptance for shipment to WIPP, whereas analytical category drums meet waste type specific decay heat requirements. The GGTP is testing test category drums at RFETS and INEEL to show compliance of drums with gas generation requirements for shipment in the TRUPACT-II and to increase the wattage limits for waste types by measurement of gas generation properties.

3.1 THE GGTP AT ROCKY FLATS ENVIRONMENTAL TECHNOLOGY SITE

At RFETS the Residue Stabilization Program determined that compliance with the wattage limit imposed by the TRUPACT-II SARP would result in an increase of 13,000 repackaged drums. As part of the GGTP, RFETS has tested drums to show compliance with TRUPACT-II gas generation requirements and to determine gas generation properties to increase the wattage limits for specific waste material types and to reduce the amount of repackaging required. (Schierloh 1998)

The method used for hydrogen gas generation testing at RFETS is referred to as the bell-jar method. The bell-jar consists of a 400-liter stainless steel overpack container called the Gas Generation Test Canister (GGTC) which hermetically encloses a vented waste drum as shown in Figure 1. Although TRUPACT-II requirements specify testing at elevated temperatures, due to safety considerations, RFETS tested drums at ambient temperatures. The GGTC collects all off-gases for a period of time and samples are taken in a SUMMA® canister. The samples are subsequently analyzed in a mass spectrometer for the concentration of hydrogen and other inorganic species.

Calculations are performed to determine the rate of hydrogen generation (moles per second). The gas generation process is modeled as a second order differential equation that is solved for gas generation rates. The calculation models the vented waste drum/GGTC system

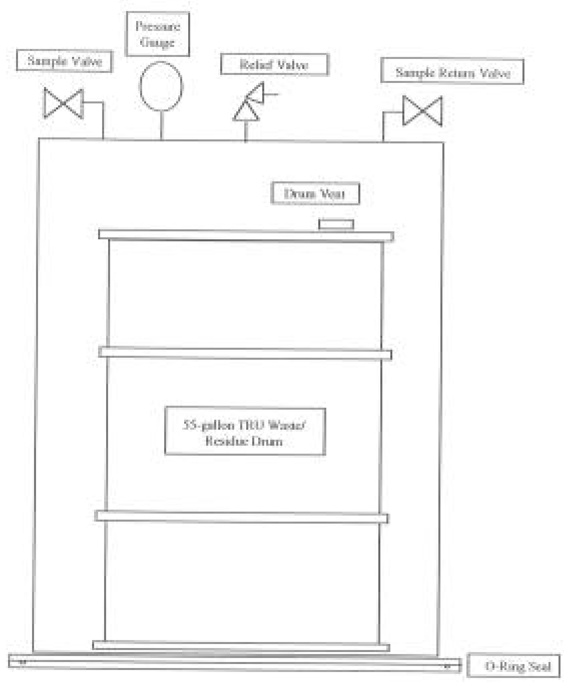

Figure 1. RFETS Bell Jar Test Apparatus

like a direct current electrical circuit with the radiolytic generation of hydrogen as the current source, the diffusion resistance of the package as the resistor, and the drum void volume and GGTC void volume as capacitors. Parameters which are experimentally established include: test duration, drum void volume, initial and final GGTC void space hydrogen concentration, and pressure and temperature of the system. (Schierloh 1998) The mathematical models used for the calculations are detailed in two reports *Gas Generation Testing -- Interim Technical Report for FY 96* (Schierloh 1996) and *Mathematical Modeling of the Build-up of Hydrogen in Radiolytic Drums* (Schierloh 1995).

Calculations are also performed in Schierloh (1998) to determine the number of hydrogen molecules generated in a material per 100 eV ionizing radiation released. The value is referred to as the $G_{effective}$ of a material. It is statistically determined by dividing the number of molecules of hydrogen generated in a drum by the amount of decay heat energy released in the drum. Note that historically, the $G(H_2)$ value of a material and decay type is defined as the number of hydrogen molecules generated in a material per 100 eV ionizing radiation *absorbe*d in the target material. In the work performed by Schierloh and others in the GGTP and MDP the G value calculated uses the total decay heat and does not attempt to determine an absolute G value for the material type by accounting for unabsorbed energy. The resulting G value is an effective G value.

The TRUPACT-II SARP uses bounding worst case G values to determine the wattage limits for organic waste materials and inorganic materials wrapped in plastic. In Schierloh (1998) after testing 63 organic residue drums, the average $G_{effective}$ value was determined to be approximately an order of magnitude less than that applied in the TRUPACT-II SARP for organic waste materials (0.346 versus 3.4). After testing 12 drums, the $G_{effective}$ was calculated to be more than an order of magnitude less for solid inorganic materials packaged in plastic (0.043 versus 1.7).

The results of 81 drums tested showed that the hydrogen generation rate in organic matrix waste drums was approximately 10% of the value expected if TRUPACT-II G values were used and the generation rate in inorganic matrix waste drums was 2.1% of predicted. The testing results allow RFETS to request an increase in the wattage limit for these two material types and reduce shipping and repackaging costs by an estimated 12 million dollars (Schierloh 1998).

3.2 THE GGTP AT IDAHO NATIONAL ENGINEERING AND ENVIRONMENTAL LABORATORY

INEEL has approximately 65,000 retrievable stored drums of which 40% to 45% are expected to fail the TRUPACT-II SAR wattage limits. The GGTP is testing drums with the objective of increasing the wattage limits for a population of drums to facilitate shipment in the TRUPACT-II and in support of the MDP.

At INEEL the GGTP has tested 114 drums as of August 1998 in a bell-jar type system attached to a continuous flow sampling and data recording system. The system is based on the TRUPACT-II SARP testing procedure outlined in Appendix 1.3.7 of the SARP. Ten bell jar style enclosures are attached to flowmeters and sample ports at the ends of stainless steel offgas lines. The bell jars are wrapped in heat tape and maintained at 135 F to 146 F to simulate worst-case thermal conditions expected in the TRUPACT-II. A computerized mass spectrometer continuously records hydrogen gas concentration. The sampling program at INEEL has encountered numerous problems due to the low offgas flow rates from the drums and the fluctuations in the system caused by barometric and atmospheric conditions.

INEEL calculations for the hydrogen generation rate in waste drums is outlined as follows: The void volume of the waste drum is determined by measuring the inflow to the drum versus temperature change during cool down at the end of the test sequence. The hydrogen generation rate is calculated from numerical integration of hydrogen flow over time, corrected for the change in hydrogen concentration in the void volume. (Edinborough 1998)

Of the 114 drums tested, 83 drums have been evaluated. Fifty-four of the evaluated drums passed the hydrogen gas generation limits, however 29 failed due to total gas generation rates. The total gas generation rates recorded for these drums may be unduly affected by atmospheric conditions and INEEL is continuing to study the problem. The hydrogen gas generation rate for approximately 50 of the drums was below the detection limit. Of the drums with detectable hydrogen gas concentrations the values are scattered such that statistically meaningful G values can not be ascertained for any specific waste material form.

3.3 THE MATRIX DEPLETION PROGRAM AT LOS ALAMOS NATIONAL LABORATORY

The purpose of the MDP is ultimately to increase the payload allowed in the TRUPACT-II by increasing the allowable wattage limits which are based on the hydrogen gas generation potential of waste materials. To further this goal, the MDP is investigating the dose dependence of G values and the phenomenon of matrix depletion. Matrix depletion is the reduction in hydrogen gas generation due to the time dependent affects of ionizing radiation on the target material. The matrix of a material that is bombarded by ionizing radiation undergoes decomposition or reaction that reduces the hydrogen generation rate with time. The phenomenon has been documented previously in Zerwekh (1979), Kosiewicz (1980, 1981), Marshall et al. (1994), and Smith et al. (1997).

The test design at Los Alamos National Laboratory (LANL) consists of 60 stainless steel cylinders, some at 60 C and some at ambient temperatures, with target materials of envirostone, polyethylene, polyvinyl chloride, and cellulose (wet and dry). The target material is sprinkled with ^{238}Pu or ^{293}Pu, folded up like a burrito, and placed in test cylinders. Averages of three replicates are reported and used for G-value calculations. Agitation of specified test cylinders to simulate transportation environments will be performed. (Benchmark 1997)

To date no published results for the MDP experimental work at LANL are available. At the July 1998 TRUPACT-II Gas Generation Test Program Annual Meeting in Albuquerque, New Mexico, Gene Mroz of LANL reported on progress in the program. In the tests conducted, temperature and isotope appeared to have no affect on matrix depletion. A reduction in G values occurred in all samples after 0.006 watt-years. The preliminary mean G-values reported were 0.14 for polyvinyl chloride with a standard deviation of 0.02 and 0.48 for wet cellulose with a standard deviation of 0.37. These values are both substantially lower than the TRUPACT-II SARP values. The testing being conducted at LANL is the most extensive research effort to date on G values for plutonium isotopes. No final published information is currently available on the results of the LANL MDP experimental work, however, it is hoped that they will result in an increase in wattage limits by approximately a factor of three which will yield positive results for the expansion of the TRUPACT-II payload.

4.0 SAVANNAH RIVER TECHNOLOGY CENTER (SRTC) RADIOLYSIS MODEL AND EXPERIMENTAL RESULTS

Experiments and modeling to predict pressure increases due to radiolysis are underway at SRTC in support of revision to the Certificate of Compliance for the 9975 packaging system. This work is being conducted by Neal Askew and Ron Livingston to authorize the transport of RFETS sand, slag, and crucible (SS&C) and fluoride residues. The approach uses a simple model to predict the pressure due to radiolysis, including the contribution from oxygen. The model distributes the alpha energy throughout the package contents based on relative mass and stopping power of the materials. The model ignores effects of self-shielding and energy transfer.

The energy deposited in water present in the payload leads to the formation of hydrogen and oxygen with an assumed $G(H_2)$ of 1.6 and a $G(O_2)$ of 0.8. Recombination and effects of the substrate are neglected. A simple Euler's method is used to calculate the depletion of the water and the accumulation of hydrogen and oxygen with time. This approach accounts for the decrease in radiolysis as the water is depleted.

Experiments are being conducted to check the conservatism of the model. Pressure measurements are not sufficient to quantify the hydrogen generation rates because of radiolytic reactions that produce or consume gas. Moisture content in the matrix is difficult to measure. The composition of gases produced by radiolysis varies dramatically. For example, nitrogen is consumed to form N_2O and NO_x, and CO_2 is produced rapidly if carbon is present. For "pure" materials, the model provides a conservative gas generation estimate.

Other observations include that the rate of pressure increase drops off more rapidly than can be attributed to water depletion and that some compounds considered inert (e. g., MgO) may enhance gas generation. Proposed future areas of study include the effects of recombination, energy transfer, and absorbed moisture on gas generation.

5.0 HYDROGEN GETTERS PROJECT

Gene Mroz is conducting the Hydrogen Getters Project at LANL. The purpose of the project is to test the response of the hydrogen getter material, DEB, in the presence of gaseous compounds commonly found in the headspace of TRU waste drums. Hydrogen getters, in the presence of catalysts such as palladium (Pd), irreversibly chemically scavenge hydrogen from the gas phase and bind it in the solid state. DEB when mixed with 25 percent carbon catalyst (5 percent Pd on carbon) has been shown to reliably and irreversibly scavenge hydrogen. In the presence of oxygen, DEB acts as a recombiner producing water. The intent of the project is to study the effect of the presence of gaseous compounds, such as VOCs, on the efficiency of the getter. (Mroz 1998)

In the Gas Getter Project, mixtures of hydrogen with other gases are passed through getter material under conditions of controlled temperature and flow rate. Hydrogen concentration is plotted versus time and compared to the hydrogen control test case. In the Gas Getter Project, gases were divided into the following functional groups and representative compounds:

chlorinated olefins	tetrachloroethylene (+), trichloroethylene (+)
chlorinated alkanes	carbon tetrachloride (+), 1,1,1-trichloroethane (+), chloroform(+) methylchloride (-)
alcohols	methanol (-)
ketones	acetone (-)
aromatics	toluene (-)
inorganic gases	CO (+)

Preliminary unpublished results presented by Gene Mroz at the TRUPACT-II Gas Generation Test Program Meeting in Albuquerque in July, 1998 showed that the compounds listed above marked with a (+) act to dampen the hydrogen scavenging abilities of DEB and those with (-) were not poisons. In general, chlorinated VOCs and carbon monoxide poisoned the getter material. A path forward for the Gas Getter Project is to develop a getter packaging that mitigates against the poisons in the form of a selective hydrogen permeable material. In addition, alternative catalyst formulations such as Pd-transition metal catalysts may reduce poisoning.

6.0 LOWER FLAMMABILITY LIMITS OF GAS MIXTURES

The TRUPACT-II SARP not only sets limits for hydrogen gas generation by use of wattage limits, but also sets limits for volatile organic compound (VOC) concentrations. The Flammability Assessment Methodology Program (FAMP) investigated the flammability of gas mixtures found in TRU waste containers at the National Institute for Occupational Safety and Health Pittsburgh Research Center. The program experimentally tested the gas mixture lower explosive limit (MLEL) of gases found in TRU waste. The experimental results were compared to predictions from the following calculational methods: Le Chatelier, modified Le Chatelier, group factor contribution, and the American Society for Testing and Materials CHETAH software. The group factor contribution method uses a linear regression model that utilizes the chemical group concept. It was developed using test results. (Loehr et al. 1998)

Tests were conducted on mixtures of flammable VOCs, nonflammable VOCs, and hydrogen gas. Four VOCs were tested in the presence and absence of hydrogen. These were 1,2-dichloroethane (DCA) to represent flammable chlorinated hydrocarbons and alkanes; 2-butanone (MEK) to represent oxygenated hydrocarbons and ketones; toluene to represent aromatic hydrocarbons; and carbon tetrachloride to represent nonflammable VOCs. (Liekhus et al. 1998)

The group factor contribution method showed better agreement with experimental results than the Le Chatelier method. The group factor contribution method was used to establish recommended limits for flammable gas VOC and flammable gas mixtures which are higher than the currently allowed TRUPACT-II SARP limit of 500 ppmv VOCs. (Loehr et al. 1998) (Liekhus et al. 1998)

7.0 PLUTONIUM METAL AND OXIDES

John Haschke of LANL has experimented extensively with plutonium dioxide. His research has centered on the kinetics of chemical reactions between plutonium dioxide and water, as well as the reaction of plutonium metal with water. Some of this research is applicable to hydrogen gas generation studies because hydrogen gas is formed from these reactions via the two reactions shown below. Under some circumstances, reaction 3 will predominate, which does not produce hydrogen gas.

(1) $PuO_2 + x\ H_2O\ =\ PuO_{2+x} + x\ H_2$

(2) $Pu + 2\ H_2O\ =\ PuO_2 + 2\ H_2$

(3) $Pu + O_2\ =\ PuO_2.$

One of the conclusions from Haschke's work is that no hydrogen will be produced in a sealed package of plutonium metal in contact with water (or even humid air) until all of the oxygen within the package is depleted (Figure 2). That occurs because all hydrogen produced by reaction 2 above quickly recombines with the oxygen present in the package to form water. That recombination reaction is actually catalyzed by the plutonium metal surface. However, once all of the oxygen is consumed, hydrogen gas is quickly produced and the package could over pressurize.

The reports provided by Haschke are summarized in the following paragraphs and are provided as attachments to this report.

LA-12999-MS (Haschke and Ricketts, 1995) evaluates whether plutonium dioxide treated in accordance with criteria adopted in DOE-STD-3013 (DOE 1996) retains enough water to cause reaction 1 above to over pressurize the container. Experiments in the past showed that 1 mass percent of water adsorbed onto a plutonium dioxide surface can result in a pressure of 50 atm of hydrogen and oxygen gas in the storage vessel. However, Haschke and Ricketts (1995) documents experiments performed at LANL which showed that calcination of plutonium dioxide at 950°C effectively removes the water from the plutonium dioxide surface and helps prevent excessive readsorption of water on that surface.

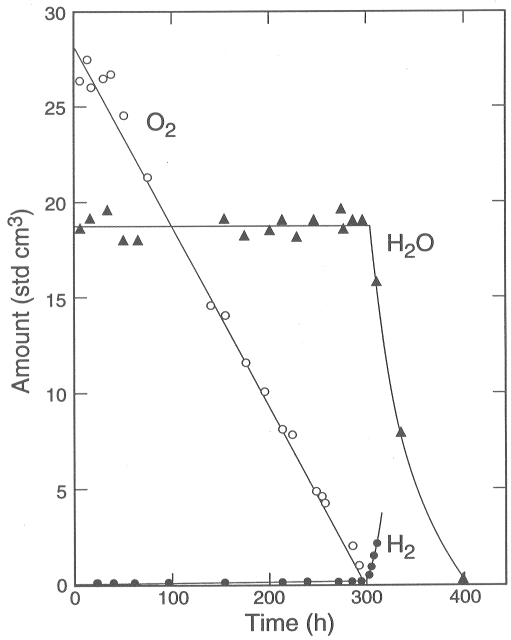

Figure 2. Plutonium Metal Reaction.

The conclusion of the above report was reaffirmed by experiments documented in LA-UR-95-2646 (Bird, et al., 1995). Those experiments showed that calcined plutonium dioxide exposed to a high humidity environment after cooling (100% relative humidity) had an equilibrium water content when removed to a lower humidity environment almost identical to a sample exposed to a low humidity environment (about 5%) immediately after calcination.

LA-13069-MS (Haschke 1995) investigates the reaction rates of uranium and plutonium metals and hydrides with water and seawater. The reaction rate of plutonium is accelerated by approximately 1000 times when exposed to seawater, while uranium reacts more slowly when exposed to seawater. Finally, this report shows that if exposure of plutonium to water is unavoidable, the reaction rate may be suppressed by making the aqueous phase basic.

Experiments and studies summarized in Haschke et al (1996) demonstrate that the corrosion rate of plutonium metal is independent of water concentration below -25°C and above 200°C, and reaction 3, shown previously, between plutonium metal and oxygen, which does not produce any hydrogen gas, predominates. However, between -25°C and 200°C, the corrosion rate is extremely sensitive to water concentrations as low as 1 ppm and reaction 2 predominates which produces hydrogen gas. But as discussed above, as long as oxygen is present, the plutonium oxide surface acts as a catalyst for recombination of the hydrogen and oxygen to form water.

The 1997 *Journal of Alloys and Compounds* article (Haschke et al, 1997) states the equilibrium loading of water on a plutonium dioxide surface is not significantly reduced until the atmosphere is maintained below 100 ppm of water, which can only occur in tightly controlled environments. This article states that formation of PuO_{2+x} is not a concern in the presence of oxygen gas, but in the presence of water, reaction 1 above could cause PuO_{2+x} to form and generate hydrogen. This article recommends further study of the kinetics of reaction 1.

LA-UR-97-2592 (Haschke et al., 1997) postulates mechanisms for two plutonium storage container failures. In both incidents, plutonium castings were stored in containers in close proximity to plastic barriers. Haschke postulates that radiolysis of the plastic created hydrogen gas inside the containers. That hydrogen gas caused plutonium hydride to form on the surface of the plutonium metal. Finally, the outermost containment boundary formed a slight leak, which allowed air to enter the package and caused a violent reaction between the plutonium hydride and both oxygen and nitrogen, catastrophically failing the storage containers. Storage of plutonium in accordance with DOE-STD-3013 should prevent this reaction from occurring since no plastic is in proximity to the plutonium; thus plutonium hydrides should not form. However, this reaction could be a concern for older plutonium packages.

The *Encyclopedia of Environmental Analysis and Remediation* (Haschke and Martz, 1998) article is an excellent synopsis of the issues encountered during plutonium storage. This article states that the reaction rate of reaction 1 above is 1.3 ± 0.2 nmol H_2/day-m^2 plutonium oxide surface at 100% relative humidity and room temperature. The activation energy for reaction 1 is estimated at 40 kJ/mol and the dependence of the rate on the surface concentration is unknown.

8.0 MATERIALS IDENTIFICATION AND SURVEILLANCE PROGRAM

The focus of the Materials Identification and Surveillance (MIS) Program meeting, held in August 1998 at Los Alamos, was the generation of hydrogen and other gases in stored plutonium-bearing materials, including plutonium oxide, salts, incinerator ash, graphite fines, fluorides, and combustible materials.

For FY98, the Core Technology projects underway at LANL are listed below along with the name of the lead principal investigator.

- Characterization Analysis
  - Plutonium chemical states (Kirk Veirs)
  - Plutonium-residue surface interactions/organic nitration (David Morris)

- Aqueous Processes
  - Actinide Solutions/Optical Spectroscopy (John Berg)
- Pyrochemistry
  - Thermodynamics (Mark Williamson)
  - Salt Distillation Science (Mary Neu)

Materials Technology
Corrosion (David Kolman)
Gas-Solid Chemistry (Gary Eller)

Methods of quantifying the hydrogen present in plutonium oxide are a focus of current research projects. Use of neutron moderation techniques under investigation by Lynn Foster (LANL) appears promising. This method uses polyethylene as a standard and preliminary results show that it is sufficiently sensitive to determine moisture content in sealed containers of plutonium oxide.

Kirk Veirs (LANL) also presented an acoustic resonance spectroscopy approach. This method is under development to provide a means of monitoring changes in gas pressure and gas composition within sealed storage containers. In the experiments, a gas cavity within a sealed container is excited using a transducer mounted on the exterior of the container. The resulting frequency response spectrum exhibits a series of peaks with position and widths dependent on the speed of sound in the gas and the resonator geometry. The intensities vary with the gas pressure. The researchers determined that the sound velocity, a function of temperature and gas composition, can be calculated with results that correlate to direct measurements to an accuracy of 0.1% for mixtures of known gases.

The effects of contaminants (e.g., chlorides) in plutonium oxide are another topic of current research. The impact of the presence of such compounds can affect stabilization, gas generation, and characterization. Lav Tandon (LANL) has presented some observations with respect to radiolysis.

Craig Leasure (LANL) is examining methods of thermal stabilization of plutonium. Parameters that affect the results include temperature, duration, and atmosphere.

Craig Leasure (LANL) has compiled results to date of the evaluations of Pu oxide cans from Rocky Flats and Hanford. The Materials Identification and Surveillance (MIS) program is analyzing 9 items from Hanford and 24 items from Rocky Flats to develop standards, site-specific stabilization plans, and to fill gaps in the information database. In FY99, 10 items from the Savannah River Site will be analyzed. Data collected include plutonium content, density, mass loss under calcination at various temperatures, and loss-on-ignition (LOI). Preliminary observations include the effect of humidity on the quantity of water taken up by the plutonium oxide. Measurement techniques for this variable are being reevaluated.

Specific MIS issues and research that may be relevant to the gas generation NUREG are as follows: Validity of the LOI method for determining the gas-generating and corrosive potential of impure plutonium bearing materials is being evaluated. LOI is the mass loss measured when a representative thermally stabilized sample of plutonium-bearing oxide is heated to confirm the elimination of residual moisture and other volatile species from the thermally stabilized material. Craig Leasure (LANL) is a suitable point of contact for this issue. Issues include the effect of volatile substances that are affected by high temperatures, yet will not contribute to gas generation during storage. The presence of uranium oxides can result in weight gain or loss during a test, yielding erroneous results.

Craig Leasure (LANL) has submitted a paper to the Journal of Nuclear Materials entitled “Conditions and Results from Thermal Stabilization of Pure and Impure Plutonium Oxides for Long-Term Storage at Department of Energy Sites. The experiments discussed in the paper lead to some conclusions regarding a reasonable temperature and duration for thermal stabilization as well as identification of suitable measuring methods.

Craig Leasure's paper (see above) also proposes a modification to the DOE-STD -3013 pressure equation which accounts for the recombination of hydrogen and oxygen that are generated by radiolysis.

9.0 SUMMARY

Research currently being conducted in the DOE community on hydrogen and other flammable gases is far reaching in scope. The largest effort is focusing on preparing an application for NRC review with the object of increasing the payload allowed in CH TRU drums to be transported in the TRUPACT-II. Such efforts include the Matrix Depletion Program; the Gas Generation Test Program, the Gas Getter Program, and the mixture lower flammability limit experiments. In addition, hydrogen generation in plutonium and plutonium-bearing materials is being addressed at LANL. The majority of the experimental work is in progress and will be difficult to incorporate into a hydrogen gas generation transportation and storage NUREG until the completion of the research programs. In addition, evaluation of G values has shown that currently accepted G values may be unduly conservative. The NUREG may, therefore, need to focus on hydrogen gas generation transportation and packaging requirements rather than defining the methodology, assumptions, and parameters that are required to show compliance.

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